

**RADIONUCLIDES IN
LICHEN, REINDEER AND MAN**

**Long-term variations and internal distribution
studied by gamma-spectrometric methods**

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RADIONUCLIDES IN LICHEN, REINDEER AND MAN

Long-term variations and internal distribution
studied by gamma-spectrometric methods

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This thesis is based on the following papers:

- I ^{137}Cs in the reindeer lichen Cladonia alpestris;
deposition, retention and internal distribution,
1961-1970.
Report LURI 1972-08 (1972)
(To be published)
- II ^{155}Eu , ^{144}Ce , ^{125}Sb , ^{106}Ru , ^{95}Zr , ^{54}Mn and ^7Be
in the reindeer lichen Cladonia alpestris;
deposition, retention and internal distribution,
1961-1970.
Report LURI 1972-09 (1972)
(To be published)
- III Sodium-22 in the food-chain: lichen-reindeer-man.
Health Physics 23, 223-230 (1972)

These papers will be referred to in the text by the Roman numerals given above.

Preliminary reports were given at:

1. Fourth Symposium on Radioactivity Investigations in Scandinavia, Oslo, Norway, October, 1967.
2. Fifth Symposium on Radioactivity Investigations in Scandinavia, Helsinki, Finland, May, 1969.
3. International Symposium on Radioecology Applied to the Protection of Man and His Environment, Rome, Italy, September, 1971.

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Radionuclides are introduced in our environment not only through emanation of gases from naturally radioactive elements in the earth's crust, and through nuclear reactions in the atmosphere caused by cosmic radiation, but also as a result of nuclear weapons tests and power production by nuclear fission.

In Table 1 are listed the origins of and relevant physical data for the radionuclides which contribute to the global radioactive contamination.

Fallout from nuclear weapons tests in the atmosphere is still the most important radioactive contamination of our environment for which man is responsible. The radionuclides, which were injected into the stratosphere by the high-yield atmospheric bomb tests carried out mainly during 1956-1958 and 1961-1962, are now almost completely deposited on the earth's surface. The majority of the undecayed radionuclides from these tests are today present in soil, water, plants, animals and man. Due to four nuclear weapons tests carried out in the atmosphere of the northern hemisphere in 1967-1970, the deposition rate of long-lived bomb-produced radionuclides has been approximately constant since 1967, at a level of about one-tenth of the maximum deposition rate in 1963-1964.

The magnitude of the deposition at a given locality varies considerably according to latitude and the amount of precipitation (UNSCEAR, 1962). The fallout over arctic and subarctic areas is, for example, considerably less than over temperate areas of the northern hemisphere. The accumulation of deposited fallout elements are, however, remarkably effective in arctic and subarctic flora, especially in lichen, which constitutes the major food-component of reindeer and caribou during winter. Reindeer- and caribou-meat are furthermore a very important food-stuff of the populations of arctic and subarctic regions. Due to these purely ecological factors, the presence of

TABLE 1

Radionuclides which contribute to the global radioactive contamination. w = nuclear weapons; r = reactors; s = power sources (space technology); n = naturally occurring.

Radio-nuclide	Type of decay	Half-life	Source
^3H	β^-	12.4 a	wr n
x ^7Be	EC, γ	53.2 d	n
^{14}C	β^-	5730 a	wr n
x ^{22}Na	β^+ , γ	2.60 a	w n
(x) ^{40}K	β^- ,EC, γ	$1.26 \cdot 10^9$ a	n
x ^{54}Mn	EC, γ	313 d	w
^{55}Fe	EC	2.7 a	w
^{85}Kr	β^- (γ)	10.8 a	wr
^{87}Rb	β^-	$4.8 \cdot 10^{10}$ a	n
^{89}Sr	β^-	52.7 d	w
^{90}Sr	β^-	28.0 a	w
$\downarrow$ ^{90}Y	β^-	64.4 h	
^{91}Y	β^- (γ)	58 d	w
x ^{95}Zr	β^- , γ	64.8 d	w
$\downarrow$ x ^{95}Nb	β^- , γ	35 d	
(x) ^{103}Ru	β^- , γ	39.6 d	w
x ^{106}Ru	β^-	367 d	w
$\downarrow$ x ^{106}Rh	β^- , γ	30 s	
x ^{125}Sb	β^- , γ	2.7 a	w
^{129}I	β^- , γ	$1.6 \cdot 10^7$ a	wr
^{131}I	β^- , γ	8.05 d	w

(continued)

TABLE 1 (continued)

Radio-nuclide	Type of decay	Half-life	Source
^{134}Cs	β^- , γ	2.1 a	w
x ^{137}Cs	β^- , γ	30.5 a	w
^{140}Ba ↓	β^- , γ	12.8 d	w
^{140}La	β^- , γ	40.2 h	
(x) ^{141}Ce	β^- , γ	32.5 d	w
x ^{144}Ce ↓	β^- , γ	284 d	w
x ^{144}Pr	β^- , γ	17.3 min	
^{147}Nd ↓	β^- , γ	11.1 d	w
^{147}Pm	β^-	2.6 a	
x ^{155}Eu	β^- , γ	1.8 a	w
(x) ^{210}Pb ↓	α , β^- , γ	21 a	n
^{210}Po	α	138 d	n
^{220}Rn	α	55 s	n
^{222}Rn	α	3.82 d	n
(x) ^{226}Ra	α , γ	1622 a	n
^{228}Ra	β^-	5.8 a	n
(x) ^{232}Th	α , γ	$1.41 \cdot 10^{10}$ a	n
^{235}U	α , γ	$7.13 \cdot 10^8$ a	n
^{238}U	α	$4.5 \cdot 10^9$ a	n
^{238}Pu	α	86.4 a	w s
^{239}Pu	α	$2.44 \cdot 10^4$ a	w
^{240}Pu	α	$6.58 \cdot 10^3$ a	w

x - radionuclide quantitatively studied in this work
 (x)- radionuclide observed but not studied in this work

radioactive elements in the environment may play a considerably more important rôle in arctic and sub-arctic areas than in most other areas of the earth. The discovery of the enhanced ^{137}Cs -level in Swedish Lapps (LIDEN, 1961) started an extensive investigation of the artificial and natural radiation environment of this population group. Similar investigations were started in Alaska, Canada, Finland, Greenland, Norway and the USSR (HANSON, 1967; HANSON et al, 1967; MIETTINEN, 1967; NEVSTRUEVA et al, 1967).

The study of the rôle of lichens as the first step in the food-chain: lichen-reindeer-man later on also developed into an investigation of the possible use of lichen as a biointegrator for radioactive fallout.

2. PURPOSE OF THE PRESENT INVESTIGATION

The present investigation should be viewed in connection with other studies on the contamination of the food-chain: lichen-reindeer-man which have been carried out at this institute. The transport of ^{137}Cs in the food-chain has been investigated (SVENSSON and LIDÉN, 1965b; LIDÉN and GUSTAFSSON, 1967; GUSTAFSSON, 1969) as well as the transport of ^{55}Fe , ^{90}Sr , ^{134}Cs , ^{210}Pb - ^{210}Po and stable lead (PERSSON, 1968, 1969, 1970a, 1970b, 1971, 1972).

If one considers the sequence: atmospheric deposition-lichen-reindeer-man, the longest mean residence-time of most radionuclides is in lichen. Therefore it is important to carefully determine the radionuclide-content in the lichen-carpet, the rate at which these radionuclides are eliminated, and the internal distribution of the radionuclides in the lichen-carpet. The present investigation was related to the lichen-carpets in the surroundings of Lake Rogen (62.3°N , 12.4°E) in the western part of the province of Härjedalen, Sweden.

The purpose of the first part of the investigation was to study the retention and distribution of ^{144}Ce , ^{137}Cs , ^{125}Sb , ^{106}Ru , ^{95}Zr and ^{54}Mn in carpets of Cladonia alpestris, which is the most frequent lichen covering the ground in this area. During the course of this work significant amounts of the radionuclides ^{155}Eu and ^7Be were also found in the lichen-carpet, and these radionuclides were also included in the study.

The second part of the investigation consists of a study of the transport of the relatively rare radionuclide ^{22}Na through the whole food-chain. In this part of the investigation, tissue-samples of reindeer which had been grazing in the Rogen area were also studied. Whole-body counting on members of the Lapp communities Mittådalen and Tännäs for ^{137}Cs -determination has been performed annually by LIDEN and co-workers. Recorded pulse height distributions have also been analysed for ^{22}Na .

3. MEASUREMENTS

The determination of low-level radioactivity in biospheric materials by radiochemical separation may in case of a complex matrix be quite difficult and time-consuming. However, by using γ -spectrometric methods it is possible to determine several radionuclides without destroying the samples. The different radionuclides reported in this investigation have all been studied in this way.

The main problem connected with the determination of small amounts of γ -emitting radionuclides in complex radionuclide mixtures, such as biospheric material, is to get high counting efficiency and good energy resolution. By means of a large NaI(Tl)-detector, the requirement on high γ -registration efficiency is well satisfied for the photon-energies of interest, and the energy resolution is often sufficient when only a limited number of γ -emitting radionuclides are present in the sample (I). In combination

with computer analysis of the pulse height distributions a NaI(Tl)-detector can also be used for studies of more complex mixtures of radionuclides, if the components are known (I,II). Using special coincidence arrangements between two or more detectors, it is possible to get better identification and quantitative determination of radionuclides with special decay schemes than when using a single detector (III). The use of large volume Ge(Li)-detectors with high energy resolution has made possible a simultaneous identification and registration of different radionuclides which have earlier not been possible to analyse without chemical separation processes (II).

4. RESULTS AND DISCUSSION

4.1 ^{137}Cs in lichen

From the standpoint of radiological health, ^{137}Cs is the most important artificially produced radionuclide found in Lapps (GUSTAFSSON, 1969; PERSSON, 1970 a). Because of the relatively short mean residence time of bomb-produced ^{137}Cs in the stratosphere (~ 1 a), the troposphere (~ 30 d), in reindeer (~ 50 d) and in man (~ 100 d) (UNSCEAR, 1972; EKMAN and GREITZ, 1967; NAVERSTEN and LIDEN, 1964), the level of ^{137}Cs in man is principally due to the retention of the radionuclide in the lichen-carpet.

Paper I presents a long-term study of the retention of ^{137}Cs in thick carpets of the reindeer lichen Cladonia alpestris. The results show a very effective retention of the radionuclide. In 1970 around 85% of all deposited ^{137}Cs was still present in a lichen-carpet having a biomass of 2 kg dry weight per m^2 , and around 60% was present in the top layer consumed by reindeer. The elimination of ^{137}Cs from a lichen-carpet is too complex to be described by means of first-order kinetics. However, as a rough approximation it is possible to assume a first-order elimination during shorter periods. During 1968-

1970 the elimination of ^{137}Cs was characterized by an effective half-time of 8-14 years for undisturbed carpets of Cladonia alpestris (2 kg m^{-2} , dry weight) studied in the Rogen area. The continuous growth and death occurring in the lichen-carpet constitute, together with the physical decay of the radionuclide, the dominating elimination process.

Samples of the lichen-carpet have been carefully fractionated in the field to enable the study of the vertical distribution of ^{137}Cs in the undisturbed lichen-carpet and in the underlying gelatinous matter, humus and soil. The ^{137}Cs -concentration in dry matter, c (nCi kg^{-1}), at the dry mass depth z (kg m^{-2}) can be expressed as

$$c = \alpha e^{-\beta z^{0.75}} \dots\dots\dots (1)$$

By means of the constants α and β it is possible to get a simple description of the vertical distribution of ^{137}Cs at different sampling-stations and -times. The results in paper I also seem to indicate that some ^{137}Cs follows the upward transport of water during periods when precipitation is low and evaporation is high, and that this ^{137}Cs is washed down during periods of high precipitation. The $^{137}\text{Cs}/\text{K}$ -ratio reaches its highest value at a depth of around 1 kg m^{-2} . The result may, as discussed in paper I, indicate that a certain amount of ^{137}Cs is not, in the same way as the K^+ -ions, available for transport in the lichen-carpet. One possible explanation is that some ^{137}Cs is still bound in undissolved radioactive fallout particles. Autoradiographic techniques have recently strongly indicated the presence of radioactive particles in the lichen-samples collected in 1970, apparently emanating from nuclear explosions (MATTSSON and SISEFSKY, 1972).

Several γ -emitting radionuclides other than ^{137}Cs contribute to the global radioactive contamination of the earth (compare with Table 1). The presence of ^{155}Eu in terrestrial biota was for the first time reported in lichen and moss (LIPPERT, 1966; MATTSSON, 1967). In earlier γ -spectrometric studies of biospheric samples the full energy peaks from this radionuclide could not be separated from the peaks from ^{144}Ce due to the insufficient energy resolution of the NaI(Tl)-spectrometers which were used. Previously reported results for ^{144}Ce from such measurements are probably seriously influenced by ^{155}Eu and thus very uncertain. Paper II presents a long-term study of ^{155}Eu and ^{144}Ce in the lichen-carpet carried out by means of high energy resolution Ge(Li)-detectors. Regarding the ^{125}Sb -content in lichen, the data found in the literature is very scanty. Paper II gives such data as well as data for ^{106}Ru and ^{54}Mn during the period 1962-1970. The presence of ^{95}Zr from more recent nuclear weapons tests was studied in 1969 and 1970. In 1970 the presence of the cosmic ray produced radionuclide ^7Be in the lichen-carpet was also studied.

All radionuclides studied in this work are more or less effectively retained in the lichen-carpet. Each radionuclide penetrates the lichen-carpet in its own special way. Using the concept "relative penetration", defined in papers I and II, it is possible to compare the distributions of two radionuclides, which can be described by means of Equation 1. According to their ability to penetrate the lichen-carpet, the different radionuclides can be arranged in the following order:

$$^{144}\text{Ce} \sim ^7\text{Be} < ^{95}\text{Zr} < ^{137}\text{Cs} < ^{106}\text{Ru} < ^{155}\text{Eu} \ll ^{125}\text{Sb}.$$

The radionuclide ^{90}Sr (PERSSON, 1971) shows a much greater depth-penetration than all these seven radionuclides. The continuously deposited naturally occurring radionuclide ^{210}Pb (PERSSON and HOLM, 1972) shows a somewhat

greater penetration than ^{155}Eu , but considerably less than ^{125}Sb .

The mean life-time of the Cladonia alpestris-carpet in the Rogen area has been estimated to be around 15 years (SKUNCKE, 1963). For the 15-year period of highest radioactive contamination (1961-1975) the absorbed dose in lichen-tissue is estimated to be 15 rad, with the dominating contributions coming from the artificially produced ^{144}Ce - ^{144}Pr ($\sim 40\%$) and from naturally occurring ^{210}Pb - ^{210}Po ($\sim 30\%$). The increased absorbed dose due to artificially produced radionuclides is not expected to produce any significant short-time biological effects on the lichen.

The absorbed dose in air at 1 m above the lichen-carpet, which was due to artificial fallout, was around 80 mrad during the period 1962-1968.

4.3 The use of lichen-carpets as biological indicators of radioactive fallout

The deposition of radionuclides on the ground is normally calculated as the sum of wet and dry deposition. The amount of dry deposition is, however, difficult to estimate because of an incomplete collection of aerosol particles. More relevant data on the total deposition is obtained from measurements on vegetation and surface soil in undisturbed horizontal areas. The use of lichen and moss as total integrating fallout-meters was suggested by SVENSSON and LIDEN (1965a) who found an almost complete retention of deposited ^{95}Zr - ^{95}Nb and ^{140}Ba - ^{140}La in a moss-carpet (Pleurozium schreberi). During recent years different species of moss have also been used as integrators in an intensive study of the deposition of stable heavy metals over Scandinavia (RÜHLING and TYLER, 1971).

In order to determine carefully the total activity of a fall-

out radionuclide per unit area of ground it is necessary to extend the sample-collection to layers lying so deep that their activity is insignificant in comparison to the activity of the overlying layers. For radionuclides, the vertical distribution of which can be described by Equation 1, it is also possible to calculate the total activity per unit area, T , according to the following equation:

$$T = T_0 + \alpha \int_{d_1}^{d_2} e^{-\beta z^{0.75}} dz \quad (\text{nCi m}^{-2}) \dots\dots\dots (2)$$

where T_0 is the area content measured in a lichen-carpet of the thickness d_1 (kg m^{-2}). The integration is extended to such a depth, d_2 (kg m^{-2}), that the activity present below d_2 is insignificant. Expressed in percent of the total activity, T , per unit area of ground for each radionuclide, the content, T_0 , retained in the lichen-carpet ($2 \text{ kg dry weight per m}^2$) was in September, 1970:

^{144}Ce - 97%, ^{95}Zr - 94%, ^{137}Cs - 86%, ^{106}Ru - 78%,
 ^{155}Eu - 71% and ^{125}Sb - 35%.

4.4 The food-chain: lichen-reindeer-man

The radionuclides which from the standpoint of man's radiological health have been devoted most interest in connection with the food-chain: lichen-reindeer-man are the artificially produced ^{137}Cs , ^{90}Sr , ^{55}Fe and the naturally occurring ^{210}Pb and ^{210}Po . Results concerning these radionuclides have earlier been published by this institute (SVENSSON and LIDÉN, 1965b; LIDÉN and GUSTAFSSON, 1967; GUSTAFSSON, 1969; PERSSON, 1969, 1970 a, 1970 b, 1971, 1972). Most of the γ -emitting radionuclides, which have been studied in paper II, are only to a very small degree transferred to the next trophic level. The internal irradiation of man by these radionuclides is insignificant in comparison with the ones mentioned above.

During the passage of lichen through the alimentary tract of lichen-grazing reindeer the gut is irradiated. The mean

life-time of a reindeer is about 7 years. During the time of highest irradiation, 1962-1968, the accumulated absorbed dose in the reindeer gut was around 3 rad, with the main contributions coming from ^{144}Ce - ^{144}Pr ($\sim 55\%$), ^{210}Pb - ^{210}Po ($\sim 20\%$) and ^{106}Ru - ^{106}Rh ($\sim 10\%$). The results of this work shows that radiation from radionuclides which pass unabsorbed through the alimentary tract of the reindeer must not be neglected in comparison with radionuclides which are absorbed by the gastro-intestinal tract and transferred to and retained in different organs of the animal.

The ^{22}Na -studies in paper III were prompted by whole-body measurements on a small number of Eskimos in Alaska, which revealed an average body-burden of 12 nCi ^{22}Na (PALMER et al, 1965), an unexpectedly high value. The present investigation has shown that the ^{22}Na -burden of Swedish Lapps was around 5-10 times lower than that of Alaskan Eskimos in spite of about the same ^{22}Na -intake for the two population groups. In paper III the difference is explained by a much lower intake of NaCl, and thus a longer mean residence time of sodium in Eskimos than in Lapps. The estimated cumulative absorbed dose in Swedish Lapps from ^{22}Na up to the year 2000 is only about 1 mrad. The accumulation of ^{22}Na in the skeleton of the reindeer has been subjected to a special study. The biological half-time of ^{22}Na incorporated in reindeer bone was estimated to be about 0.5 year.

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